

09/890,086



Consommation
et Corporations Canada

Consumer and
Corporate Affairs Canada

Bureau des brevets

Patent Office

Ottawa, Canada
K1A 0C9

(21) (A1)	2,073,135
(22)	1992/07/03
(43)	1993/01/06

5,025,3/79

(51) INTL.CL.⁵ C07D-401/12; C07D-401/14; C07D-413/14; A01N-047/36

(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Salts of Pyridylsulfonylureas as Herbicides and Plant Growth Regulators, Their Preparation and Their Use

(72) Kehne, Heinz - Germany (Federal Republic of) ;
Haaf, Klaus - Germany (Federal Republic of) ;
Bauer, Klaus - Germany (Federal Republic of) ;
Bieringer, Herman - Germany (Federal Republic of) ;

(73) Hoechst Aktiengesellschaft - Germany (Federal Republic of) ;

(30) (DE) P 41 22 306.3 1991/07/05

(57) 9 Claims

Notice: The specification contained herein as filed

Canada

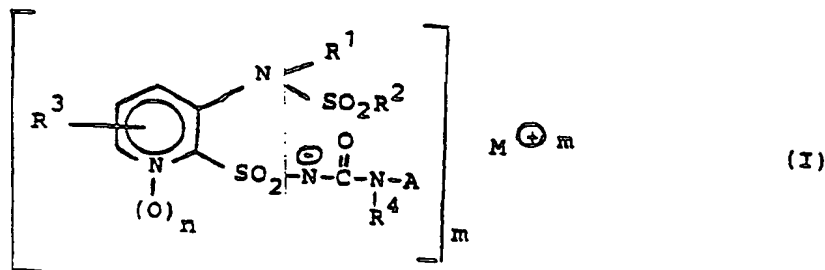
CCA 3254 (10-89) 41

2073135

ABSTRACT OF THE DISCLOSURE:

Salts of pyridylsulfonylureas as herbicides and plant growth regulators, their preparation and their use

Compounds of the formula (I)



in which

R^1 is H or (in each case substituted) alkyl, alkenyl, alkynyl, alkylsulfonyl, phenylsulfonyl, alkoxy or alkylsulfonyl,

R^2 is (substituted) alkyl, (substituted) phenyl or dialkylamino, or

R^1 and R^2 together are a (substituted) C_3 - C_4 -alkylene chain,

R^3 is H, alkyl, haloalkyl, Hal, NO_2 , CN, alkoxy, haloalkoxy, alkylthio, alkoxyalkyl, alkoxy-carbonyl, mono- or dialkylamino, alkylsulfonyl, alkylsulfinyl, (substituted) amidosulfonyl or (substituted) carboxamide,

R^4 is H or CH_3 , $n = 0$ or 1 , $m = 1$ or 2 ,

A is 4,6-disubstituted pyrimidinyl (see claim 1),

M is a monovalent or divalent cation (see claim 1),

are suitable as herbicides and plant growth regulators. They are prepared, for example, from the corresponding sulfonylurea by reaction with an alcoholate, carbonate or amine.

Description

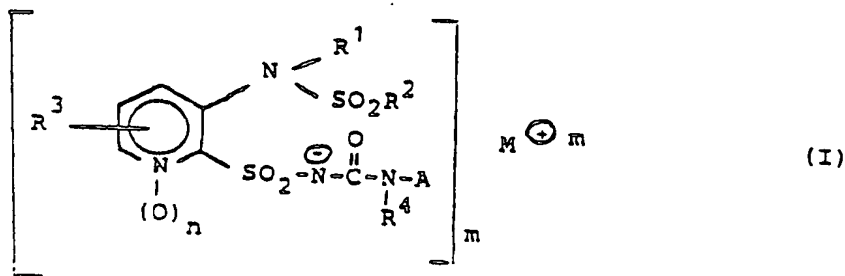
Salts of pyridylsulfonylureas as herbicides and plant growth regulators, their preparation and their use

5 It has been disclosed that some 2-pyridylsulfonylureas have herbicidal and plant-growth-regulating properties; cf. EP-A-13,480 (US-A-4,435,206), EP-A-272,855 (US-A-4,838,926), EP-A-84,224 (US-A-4,629,494), US-A-4,421 550, EP-A-103,543 (US-A-4,579,583),
 10 US-A-4,946,494, US-A-4,487,626, EP-A-125,864 (US-A-4,723,991), WO 88/04297, EP-A-178,101 (US-A-4,756,742).

2-Pyridylsulfonylureas having specific radicals in the 3-position of the pyridyl radical have already been
 15 proposed as herbicides and plant growth regulators in PCT Patent Application No. PCT/EP 90/02308 (WO-91/10660). Salts of the compounds are only mentioned in this publication as alternative use forms of the herbicides.

It has now been found that certain agriculturally utiliz-
 20 able salts of 2-pyridylsulfonylureas having specific radicals in the 3-position of the pyridyl radical are particularly suited as herbicides and growth regulators.

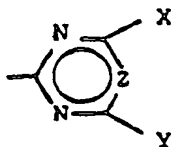
The present invention relates to compounds (salts) of the formula (I)



in which

- 5 R^1 is H, (C_1-C_6) alkyl which is unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) -alkoxy, (C_1-C_4) alkylthio, (C_1-C_4) alkylsulfinyl, (C_1-C_4) alkylsulfonyl, $[(C_1-C_4)$ alkoxy]carbonyl and CN, (C_3-C_6) alkenyl which is unsubstituted or substituted by one or more halogen atoms, (C_3-C_6) alkynyl which is unsubstituted or substituted by one or more halogen atoms, (C_1-C_4) -alkylsulfonyl which is unsubstituted or substituted by one or more halogen atoms, phenylsulfonyl where the phenyl radical is unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) alkyl and (C_1-C_4) alkoxy, (C_1-C_4) alkoxy or $[(C_1-C_4)$ alkyl]carbonyl, which is unsubstituted or substituted by one or more halogen atoms,
- 10 R^2 is (C_1-C_4) alkyl which is unsubstituted or substituted by one or more halogen atoms, phenyl, where the phenyl radical is unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) alkyl and (C_1-C_4) alkoxy, or is di- $[(C_1-C_4)$ alkyl]amino or
- 15 R^1 and R^2 together are a chain of the formula $-(CH_2)_o-$ where the chain can additionally be substituted by 1 to 4 (C_1-C_3) alkyl radicals, and o is 3 or 4,
- 20 R^3 is H, (C_1-C_4) alkyl, preferably (C_1-C_3) alkyl, (C_1-C_3) haloalkyl, halogen, NO_2 , CN, (C_1-C_3) -alkoxy, (C_1-C_3) haloalkoxy, (C_1-C_3) alkylthio, (C_1-C_3) alkoxy- (C_1-C_3) alkyl, $[(C_1-C_3)$ alkoxy]carbonyl, (C_1-C_3) alkylamino, di- $[(C_1-C_3)$ alkyl]-amino, (C_1-C_3) alkylsulfinyl, (C_1-C_3) alkylsulfonyl, $SO_2NR^aR^b$ or $C(O)NR^aR^b$,
- 25 R^a and R^b independently of one another are H, (C_1-C_3) -alkyl, (C_3-C_4) alkenyl or propargyl, or together are $-(CH_2)_4-$, $-(CH_2)_5-$ or $-CH_2CH_2OCH_2CH_2-$,
- 30
- 35

R^4 is H or CH_3 ,
 n is zero or 1,
 m is 1 or 2,
 A is a radical of the formula



5 X and Y independently of one another are H, halogen, (C_1-C_3) alkyl, (C_1-C_3) alkoxy or (C_1-C_3) alkylthio, the abovementioned alkyl-containing radicals being unsubstituted or mono- or polysubstituted by halogen or mono- or disubstituted by
 10 (C_1-C_3) alkoxy or (C_1-C_3) alkylthio, furthermore a radical of the formula NR^5R^6 , (C_3-C_6) cycloalkyl, (C_2-C_4) alkenyl, (C_2-C_4) alkynyl, (C_3-C_4) alkenyloxy or (C_3-C_4) alkynyloxy,
 Z is CH or N,
 15 R^5 and R^6 independently of one another are H, (C_1-C_3) alkyl or (C_3-C_4) alkenyl, and
 M is an atom selected from the group comprising the alkali metals, alkaline earth metals or a group of the formula M1



20 in which R^7 , R^8 , R^9 and R^{10} independently of one another are H, (C_1-C_{12}) alkyl, (C_3-C_6) alkenyl, (C_3-C_6) alkynyl, (C_3-C_6) cycloalkyl or phenyl, the last 5 radicals mentioned in each case independently of one another being unsubstituted or
 25 substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) alkyl, hydroxyl, (C_1-C_4) alkoxy, thio, (C_1-C_4) alkylthio, $[(C_1-C_4)$ alkoxy]carbonyl and

5 optionally substituted phenyl, or two of the radicals R^7 to R^{10} together are a saturated or unsaturated chain of 3-7 carbon atoms, it being possible for 1-2 carbon atoms to be replaced by atoms selected from the group comprising O, N or S and for the chain to be substituted by 1-3 (C_1 - C_4)alkyl radicals, and the remaining two radicals have the abovementioned meanings of individual radicals R^7 to R^{10} .

10 In formula (I) and hereinafter, alkyl, alkoxy, haloalkyl, alkylamino and alkylthio radicals and the corresponding unsaturated and/or substituted radicals can in each case be straight-chain or branched. Alkyl radicals, also in
15 composite meanings such as, inter alia, alkoxy and haloalkyl, are, for example, methyl, ethyl, n- or i-propyl or n-, i-, t- or 2-butyl; alkenyl and alkynyl radicals have the meanings of the unsaturated radicals which are possible and which correspond to the alkyl radicals, such as 2-propenyl, 2- or 3-butenyl, 2-propynyl
20 or 2- or 3-butyne. Halogen is fluorine, chlorine, bromine or iodine; haloalkyl is alkyl which is substituted by one or more atoms selected from the halogen group; haloalkyl is, for example, CF_3 , CHF_2 or CH_2CF_3 . Optionally substituted phenyl is preferably unsubstituted phenyl or phenyl which is substituted by one or more,
25 preferably 1 to 3, radicals selected from the group comprising halogen, alkyl, alkoxy, nitro, cyano, alkoxy-carbonyl, alkamoyl, carbamoyl, mono- and dialkylamino-carbonyl, mono- and dialkylamino, alkylsulfinyl or
30 alkylsulfonyl, preferred alkyl-containing radicals being those having 1 to 4 carbon atoms.

Preferred compounds of the formula I are those in which R^3 , R^a , R^b and A are as defined above and $n = \text{zero}$,
35 R^1 is H, (C_1 - C_4)alkyl which is unsubstituted or substituted by one or more halogen atoms or by a radical selected from the group comprising (C_1 - C_3)alkoxy, (C_1 - C_3)alkylthio, (C_1 - C_3)alkyl-

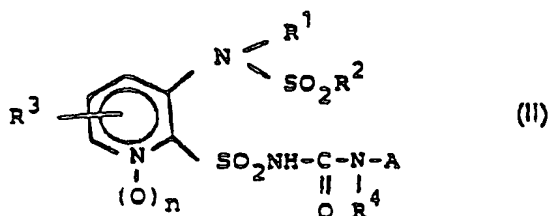
- sulfonyl, [(C₁-C₄)alkoxy]carbonyl and CN, or is (C₃-C₄)alkenyl, (C₃-C₄)alkynyl, (C₁-C₄)alkylsulfonyl, (C₁-C₃)alkoxy or [(C₁-C₄)alkyl]carbonyl, and
- 5 R² is (C₁-C₄)alkyl which is unsubstituted or substituted by 1 to 3 radicals selected from the halogen group, or
- R¹ and R² together are a chain of the formula -(CH₂)_o-, in which o is 3 or 4.
- 10 Particularly preferred compounds of the formula (I) are those in which
- R¹ is hydrogen, (C₁-C₄)alkyl or halo(C₁-C₄)alkyl,
- R² is (C₁-C₃)alkyl or halo(C₁-C₄)alkyl,
- R³ is H, (C₁-C₃)alkyl, (C₁-C₃)alkoxy, halogen or
- 15 (C₁-C₃)alkylthio,
- Z CH or N,
- X is (C₁-C₃)alkyl, (C₁-C₃)alkoxy, (C₁-C₃)haloalkyl, (C₁-C₃)haloalkoxy, (C₁-C₃)alkoxy-(C₁-C₃)alkyl or (C₁-C₃)alkoxy-(C₁-C₃)alkoxy, and
- 20 Y is halogen, (C₁-C₃)alkyl, (C₁-C₃)alkoxy or (C₁-C₃)alkylthio, it being possible for the last 3 radicals mentioned to be monosubstituted or polysubstituted by halogen or monosubstituted or disubstituted by radicals selected from
- 25 the group comprising (C₁-C₃)alkoxy and (C₁-C₃)alkylthio,
- or a radical of the formula NR⁵R⁶ in which R⁵ and R⁶ have the abovementioned meaning, or (C₃-C₆)cycloalkyl, (C₂-C₄)alkynyl, (C₃-C₄)alkenyloxy or (C₃-C₄)alkynyloxy.
- 30
- X is preferably (C₁-C₂)alkyl, (C₁-C₂)alkoxy, OCF₂H, CF₃ or OCH₂CF₃ and
- Y is preferably (C₁-C₂)alkyl, (C₁-C₂)alkoxy, halogen or OCF₂H.
- 35 M is preferably Na, Li, K, Ca or a group of the formula M₁



(M1)

in which R^7 , R^8 , R^9 and R^{10} independently of one another are H, (C_1-C_{12}) alkyl, (C_3-C_4) alkenyl, (C_3-C_4) alkynyl, (C_3-C_6) cycloalkyl, $[(C_1-C_4)$ alkoxy]carbonyl-
 5 (C_1-C_4) alkyl, hydroxy- (C_1-C_4) alkyl, phenyl, benzyl, 1-phenylethyl or 2-phenylethyl, or
 M is preferably piperidine, pyrrolidine, morpholine or pyridine.

The present invention furthermore relates to processes for the preparation of the compounds of the formula (I),
 10 which comprise reacting a compound of the formula (II)



in which A, R^1 , R^2 , R^3 , R^4 and n are defined as in formula (I) as claimed in claim 1, with a base of the formula (IIIa) or (IIIb)



in which M and m are defined as in formula (I) and
 15 R^{11} is H or an aliphatic or aromatic organic radical, preferably (C_1-C_4) alkyl, and p is the number 2 in the event that m = 1 and the number 1 in the event that m = 2,

or, in the event that $R^{10} = H$, reacting a compound of the
 20 formula (II) with an amine of the formula (IV)



in

which R^7 to R^9 are defined as in formula M1.

The reaction is carried out for example in the presence of solvents which are inert under the reaction conditions. Suitable solvents are inorganic solvents such as water or organic solvents, for example alcohols, such as, for example, methanol or ethanol, halogenated hydrocarbons such as dichloromethane, ethers such as tetrahydrofuran or dioxane, ketones such as acetone or MIBK (methyl isobutyl ketone), amides such as DMF, nitriles such as acetonitrile, and sulfoxides such as DMSO. The reaction temperatures are mostly between 0°C and the boiling point of the solvent.

An analogous process with reaction conditions is described in WO 90/06308.

The products of the formula (I) can be isolated in virtually quantitative yield and in good purity, in some cases directly by filtration or, if they are readily soluble, after removing the solvent by distillation.

The starting materials of the formula (II) are known or can be prepared analogously to known processes. Their synthesis is described, for example, in DE-A-4,000,503. Most of the bases of the formulae (IIIa) and (IIIb) are standard reagents and in some cases commercially available even on an industrial scale. They are, for example, alkali metal hydroxides, alkali metal alcoholates, alkali metal carbonates, alkaline earth metal hydroxides, alkaline earth metal alcoholates or alkaline earth metal carbonates, but also quaternary ammonium hydroxides, such as, for example, tetrabutylammonium hydroxide.

The bases of the formula (IV) comprise ammonia, primary,

secondary and tertiary amines such as, for example, methylamin , diethylamine, tri thylamine, diethanolamine or benzylamine, but also heterocyclic amines such as, for example, pyridine, pyrrolidine, piperidine or morpholine.

5 The bases of the formulae (IIIa), (IIIb) or (IV) are employed, for example, in amounts of 0.5 to 1.5 mol, preferably 0.9 to 1.1 mol per mole of sulfonylurea of the formula II.

10 The compounds of the formula (I) according to the invention have an excellent herbicidal activity against a broad range of economically important monocotyledon and dicotyledon harmful plants. The active substances also act efficiently on perennial weeds which produce shoots from rhizomes, rootstocks or other perennial organs and
15 which are difficult to control. In this context, it does not matter whether the substances are applied before sowing, pre-emergence or post-emergence. Specifically, examples may be mentioned of some representatives of the monocotyledon and dicotyledon weed flora which can be
20 controlled by the compounds according to the invention, without the enumeration being a restriction to certain species.

25 Examples of weed species on which the active substance acts efficiently are, from amongst the monocotyledons, Avena, Lolium, Alopecurus, Phalaris, Echinochloa, Digitaria, Setaria and also Cyperus species from the annual sector and from amongst the perennial species Agropyron, Cynodon, Imperata and Sorghum, and also perennial Cyperus species.

30 In the case of the dicotyledon weed species, the range of action extends to species such as, for example, Galium, Viola, Veronica, Lamium, Stellaria, Amaranthus, Sinapis, Ipomoea, Matricaria, Abutilon and Sida from amongst the annuals, and Convolvulus, Cirsium, Rumex and Artemisia in
35 the case of the perennial weeds.

5 The active substances according to the invention equally effect outstanding control of weeds which occur under the specific conditions of rice growing, such as, for example, Sagittaria, Alisma, Eleocharis, Scirpus and Cyperus.

10 If the compounds according to the invention are applied to the soil surface before germination, then the weed seedlings are either prevented completely from emerging, or the weeds grow until they have reached the cotyledon stage but then their growth stops, and, eventually, after three to four weeks have elapsed, they die completely.

15 If the active substances are applied post-emergence on the green parts of the plants, growth likewise stops drastically a very short time after the treatment and the weed plants remain at the growth stage of the point of time of application, or they die completely after a certain time, so that in this manner competition by the weeds, which is harmful to the crop plants, is eliminated at a very early point in time and in a sustained manner.

20 Even though the compounds according to the invention have an excellent herbicidal activity against monocotyledon and dicotyledon weeds, crop plants of economically important crops, such as, for example, wheat, barley, rye, rice, maize, sugar beet, cotton and soya, are
25 damaged not at all, or only to a negligible extent. For these reasons, the present compounds are highly suitable for selectively controlling undesired plant growth in plantings for agricultural use.

30 Moreover, the substances according to the invention have outstanding growth-regulatory properties in crop plants. They engage in the plant metabolism in a regulating manner and can therefore be used for influencing plant constituents and facilitating harvesting in a targeted manner, for example by triggering desiccation and stunted
35 growth. Moreover, they are also suitable for generally

controlling and inhibiting undesirable vegetative growth without destroying the plants in the process. Inhibition of vegetative growth is highly important in many monocotyledon and dicotyledon crops since lodging can thereby be reduced or prevented completely.

The compounds according to the invention can be applied in the form of wettable powders, emulsifiable concentrates, sprayable solutions, dusting agents or granules in the customary formulations. The invention therefore also relates to herbicidal and plant-growth-regulating compositions which comprise the compounds of the formula (I).

The compounds of the formula (I) can be formulated in many ways, depending on the prevailing biological and/or chemicophysical parameters. The following are examples of formulations which are possible: wettable powders (WP), water-soluble powders (SP), water-soluble concentrates, emulsifiable concentrates (EC), emulsions (EW) such as oil-in-water and water-in-oil emulsions, sprayable solutions, suspension concentrates (SC), dispersions on an oil or water basis, oil-miscible solutions, capsule suspensions (CS), dusts (DP), seed-dressing agents, granules for broadcasting and soil application, granules (GR) in the form of microgranules, spray granules, coated granules and adsorption granules, water dispersible granules (WG), water-soluble granules (SG), ULV formulations, microcapsules and waxes.

These abovementioned formulation types are described, for example in: Winnacker-Kuchler, "Chemische Technologie [Chemical Technology]", Volume 7, C. Hauser Verlag Munich, 4th Ed. 1986; Wade van Valkenburg, "Pesticides Formulations", Marcel Dekker N.Y., 1973; K. Martens, "Spray Drying Handbook", 3rd Ed. 1979, G. Goodwin Ltd. London.

The formulation auxiliaries required, such as inert

- materials, surfactants, solvents and other additives are also known and are described, for example, in: Watkins, "Handbook of Insecticid Dust Diluent and Carriers", 2nd Ed., Darland Books, Caldwell N.J., H.v. Olphen, 5 "Introduction to Clay Colloid Chemistry"; 2nd Ed., J. Wiley & Sons, N.Y.; C. Marsden, "Solvents Guide"; 2nd Ed., Interscience, N.Y. 1963; McCutcheon's "Detergents and Emulsifiers Annual", MC Publ. Corp. Ridgewood N.J.; Sisley and Wood, "Encyclopedia of Surface 10 Active Agents", Chem. Publ. Co. Inc., N.Y. 1964; Schönfeldt, "Grenzflächenaktive Äthylenoxidaddukte [Surface-active Ethylene Oxide Adducts]", Wiss. Verlagsgesell., Stuttgart 1976; Winnacker-Küchler, "Chemische Technologie [Chemical Technology]", Volume 7, 15 C. Hauser Verlag Munich, 4th Ed. 1986.

Based on these formulations, it is also possible to produce combinations with other pesticidally active substances, fertilizers and/or growth regulators, for example in the form of a readymix or a tank mix.

- 20 Wettable powders are preparations which are uniformly dispersible in water and which, besides the active substance, also contain ionic and non-ionic surfactants (wetting agents, dispersants), for example polyoxyethylated alkylphenols, polyoxyethylated fatty alcohols and 25 fatty amines, fatty alcohol polyglycol ether sulfates, alkanesulfonates or alkylbenzenesulfonates, sodium ligninsulfonate, sodium 2,2'-dinaphthylmethane-6,6'-disulfonate, sodium dibutyl-naphthalenesulfonate, or alternatively sodium oleoylmethyltaurate, in addition to a 30 diluent or inert substance.

- Emulsifiable concentrates can be prepared, for example, by dissolving the active substance in an inert organic solvent, for example butanol, cyclohexanone, dimethylformamide, xylene or also higher-boiling aromatic 35 compounds or hydrocarbons, with the addition of one or more emulsifiers. Examples of emulsifiers which can be

5 used are: calcium salts of an alkylarylsulfonic acid, such as calcium dodecylbenzenesulfonate, or non-ionic emulsifiers, such as fatty acid polyglycol esters, alkylaryl polyglycol ethers, fatty alcohol polyglycol
10 ethers, propylene oxide/ethylene oxide condensation products, alkyl ethers, sorbitan fatty acid esters, polyoxethylene sorbitan fatty acid esters or polyoxethylene sorbitol esters.

10 Dusts can be obtained by grinding the active substance with finely divided solid substances, for example talc or natural clays, such as kaolin, bentonite and pyrophyllite, or diatomaceous earth.

15 Granules can be produced either by spraying the active substance onto adsorptive, granulated inert material or by applying active substance concentrates onto the surface of carriers, such as sand, kaolinites or granulated inert materials, by means of binders, for example polyvinyl alcohol, sodium polyacrylate or, alternatively, mineral oils. Suitable active substances can also be
20 granulated in the manner which is conventional for the production of fertilizer granules, if desired in a mixture with fertilizers. Water-dispersible granules are generally prepared by the customary processes such as spray drying, fluidized-bed granulation, disk granulation, mixing with high-speed mixers, and extrusion
25 without solid inert material.

The agrochemical preparations generally comprise 0.1 to 99% by weight, in particular 0.1 to 95% by weight, of active substances of the formula (I).

30 The active substance concentration in wettable powders is, for example, about 10 to 90% by weight; the remainder to 100% by weight comprises conventional formulation components. In the case of emulsifiable concentrates, the active substance concentration can be about 1 to 85% by
35 weight, mostly 5 to 80% by weight. Formulations in the

5 form of dusts usually contain about 1 to 25% by weight, mostly 5 to 20% by weight, sprayable solutions about 0.2 to 20% by weight, mostly 2 to 20% by weight. In the case of granules, the active substance content depends partly on whether the active compound is liquid or solid. The active substance content in the case of the water-dispersible granules is generally between 10 and 90% by weight.

10 In addition, the active substance formulations mentioned comprise, if appropriate, the adhesives, wetting agents, dispersants, emulsifiers, penetrants, preservatives, antifreeze agents and solvents, fillers, colorants and carriers, defoamers, evaporation inhibitors, pH regulators and viscosity regulators which are conventional in
15 each case.

For use, the formulations, present in commercially available form, are diluted, if appropriate, in a customary manner, for example by means of water in the case of wettable powders, emulsifiable concentrates,
20 dispersions and water-dispersible granules. Preparations in the form of dusts, granules for soil application or for broadcasting as well as sprayable solutions are usually not further diluted with other inert substances before use.

25 The formulations are applied, for example, to the plants, parts of the plants, seeds of the plants (seed dressing) or the area under cultivation.

The application rate required of the compounds of the formula (I) varies as a function of the external conditions such as, inter alia, temperature, humidity, and
30 nature of the herbicide used. It can vary within wide limits, for example between 0.001 and 10.0 kg/ha or more of active ingredient, but it is preferably between 0.005 and 5 kg/ha.

If appropriate, mixtures or mixed formulations with other active substances such as, for example, insecticides, acaricides, herbicides, safeners, fertilizers, growth regulators or fungicides are also possible.

5 A. Chemical examples

- a) Sodium salt of 3-(4,6-dimethoxypyrimidin-2-yl)-1-[3-(N-methyl-N-methylsulfonylamino)pyrid-2-yl-sulfonyl]urea (Tabulated Example 1)

10 3.0 g (0.13 gram-equivalent) of sodium are dissolved in 300 ml of methanol. 58.0 g (0.13 mol) of 3-(4,6-dimethoxypyrimidin-2-yl)-1-[3-(N-methyl-N-methylsulfonylamino)pyrid-2-ylsulfonyl]urea are added to this solution at room temperature, and the mixture is subsequently refluxed for 30 minutes. The mixture is cooled, and the solvent is removed in vacuo, giving 15 60.7 g (100% of theory) of the abovementioned sodium salt of melting point 203-205°C (decomp.).

- b) Triethylammonium salt of 3-(4,6-dimethoxypyrimidin-2-yl)-1-[3-(N-methyl-N-methylsulfonylamino)pyrid-2-ylsulfonyl]urea (Tabulated Example 12)

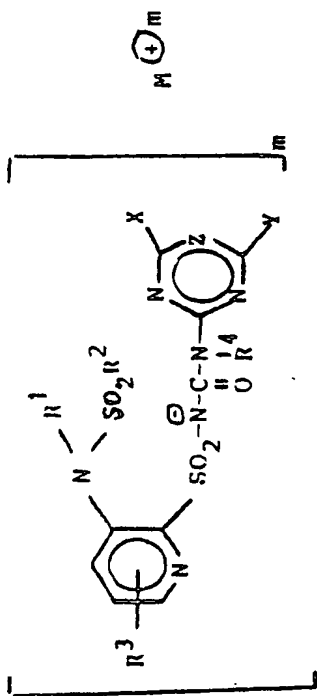
20 To a solution of 2.23 g (5 mmol) of 3-(4,6-dimethoxypyrimidin-2-yl)-1-[3-(N-methyl-N-methylsulfonylamino)pyrid-2-ylsulfonyl]urea in 50 ml of dichloromethane there is added 0.51 g (5 mmol) of triethylamine, and the mixture is refluxed for 25 30 minutes. The mixture is cooled, the solvent is removed in vacuo, and the residue is triturated with diethyl ether. This gives 2.5 g (91% of theory) of the abovementioned triethylammonium salt of melting point 99-101°C.

- 30 c) Lithium salt of 3-(4,6-dimethoxypyrimidin-2-yl)-1-[3-(N-methyl-N-methylsulfonylamino)pyrid-2-ylsulfonyl]urea (Tabulated Example 2)

5 To a solution of 0.21 g (5 mmol) of lithium
hydroxide (monohydrate) in 50 ml of methanol there
are added 2.23 g (5 mmol) of 3-(4,6-dimethoxypyri-
midin-2-yl)-1-[3-(N-methyl-N-methylsulfonylamino)-
pyrid-2-ylsulfonyl]urea, and the mixture is refluxed
for 30 minutes. The mixture is cooled, the solvent
is removed in vacuo, and the residue is triturated
with diethyl ether. This gives 2.2 g (98% of theory)
10 of the abovementioned lithium salt of melting point
247-249°C (decomp.)

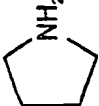
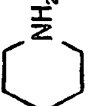
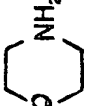
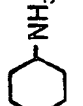
The compounds in Table 1 below are obtained analogously
to the processes of Examples 1-3.

Table 1



Compd. No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	M	m	M.P. [°C]
1	CH ₃	CH ₃	H	H	OCH ₃	OCH ₃	CH	Na	1	203-205 (decomp.)
2	"	"	"	"	"	"	"	Li	1	247-249 "
3	"	"	"	"	"	"	"	K	1	213-216 "
4	"	"	"	"	"	"	"	Mg	2	
5	"	"	"	"	"	"	"	Ca	2	
6	"	"	"	"	"	"	"	NH ₄	1	
7	"	"	"	"	"	"	"	CH ₃ NH ₂	1	

Compd. No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	M	m	M.P. [°C]
8	CH ₃	CH ₃	H	H	OCH ₃	OCH ₃	CH	(CH ₃) ₂ NH ₂	1	
9	"	"	"	"	"	"	"	(CH ₃) ₃ NH	1	
10	"	"	"	"	"	"	"	C ₂ H ₅ NH ₃	1	
11	"	"	"	"	"	"	"	(C ₂ H ₅) ₂ NH ₂	1	
12	"	"	"	"	"	"	"	(C ₂ H ₅) ₃ NH	1	99-101
13	"	"	"	"	"	"	"	C ₄ H ₉ NH ₃	1	127-129
13a	"	"	"	"	"	"	"	(C ₄ H ₉) ₄ N	1	
14	"	"	"	"	"	"	"	HOCH ₂ CH ₂ NH ₃	1	
15	"	"	"	"	"	"	"	(HOCH ₂ CH ₂) ₂ NH ₂	1	65-68
16	"	"	"	"	"	"	"	(HOCH ₂ CH ₂) ₃ NH	1	142-144
17	"	"	"	"	"	"	"	C ₁₂ H ₂₅ NH ₃	1	
18	"	"	"	"	"	"	"	C ₆ H ₅ -CH ₂ NH ₃	1	
19	"	"	"	"	"	"	"	C ₆ H ₅ -CH ₂ CH ₂ NH ₃	1	

Compd. No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	M	m	M.P. [°C]
20	CH ₃	CH ₃	H	H	OCH ₃	OCH ₃	CH	C ₆ H ₅ -NH ₃	1	
21	"	"	"	"	"	"	"	H ₃ C ₂ OCO-CH ₂ NH ₃	1	
22	"	"	"	"	"	"	"		1	
23	"	"	"	"	"	"	"		1	
24	"	"	"	"	"	"	"		1	
25	"	"	"	"	"	"	"		1	

[H]N1C=CC=CC=C1

[illegible]

Compd. No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	M	m	M.P. [°C]
51	CH ₃	CH ₃	6-CH ₃	H	OCH ₃	OCH ₃	CH	NH ₄	1	
52	"	"	"	"	"	"	"	(C ₂ H ₅) ₃ NH	1	oil
53	CH ₃	CH ₃	6-Cl	H	OCH ₃	OCH ₃	CH	Na	1	
54	"	"	6-OCH ₃	"	"	"	"	Na	1	
55	"	"	5-CH ₃	"	"	"	"	Na	1	
56	"	"	4-CH ₃	"	"	"	"	Na	1	
57	CH ₃	CH ₃	H	H	Cl	OCH ₃	CH	Na	1	
58	"	"	"	"	OCH ₃	OC ₂ H ₅	CH	Na	1	
59	"	"	"	"	OCH ₃	SCH ₃	CH	Na	1	
60	"	"	"	"	F	OCH ₃	CH	Na	1	
61	"	"	"	"	OCH ₃	OCH ₃	N	Na	1	
62	"	"	"	"	OCH ₃	CF ₃	CH	Na	1	
63	"	"	"	CH ₃	OCH ₃	CH ₃	N	Na	1	

2073135

Compd. No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	M	m	M.P. [°C]
64	CH ₃	CF ₃	H	H	OCH ₃	OCH ₃	CH	Na	1	
65	"	C ₂ H ₅	"	"	"	"	"	Na	1	
66	CHF ₂	CH ₃	"	"	"	"	"	"	1	
67	-C ₂ H ₅	"	"	"	"	"	"	"	1	
68	-CH(CH ₃) ₂	"	"	"	"	"	"	"	1	
69	-C ₃ H ₇	"	"	"	"	"	"	"	1	
70	-CH ₂ CH=CH ₂		"	"	"	"	"	"	1	
71	-CH ₂ C≡CH	"	"	"	"	"	"	"	1	
72	-CH ₂ CH ₂ Cl	"	"	"	"	"	"	"	1	
73	CH ₂ CH ₂ OCH ₃	"	"	"	"	"	"	"	1	
74	CH ₃	CH ₃	"	"	OCHF ₂	OCHF ₂	CH	"	1	

B. Formulation Examples

- 5 a) A dusting agent is obtained by mixing 10 parts by weight of a compound of the formula (I) and 90 parts by weight of talc as inert substance, and comminuting the mixture in a hammer mill.
- 10 b) A wettable powder which is readily dispersible in water is obtained by mixing 25 parts by weight of a compound of the formula (I), 64 parts by weight of kaolin-containing quartz as the inert substance, 10 parts by weight of potassium ligninsulfonate and 1 part by weight of sodium oleoylmethyltaurate as the wetting and dispersing agent, and grinding the mixture in a pinned disk mill.
- 15 c) A dispersion concentrate which is readily dispersible in water is obtained by mixing 20 parts by weight of a compound of the formula (I) with 6 parts by weight of alkylphenol polyglycol ether (^oTriton X 207), 3 parts by weight of isotridecanol polyglycol ether (8 EO) and 71 parts by weight of paraffinic mineral oil (boiling range, for example, about 255 to above 277°C), and grinding the mixture in a ball mill to a fineness of below 5 microns.
- 20 d) An emulsificable concentrate is obtained from 15 parts by weight of a compound of the formula (I), 75 parts by weight of cyclohexanone as the solvent and 10 parts by weight of oxethylated nonylphenol as the emulsifier.
- 25 e) Water-dispersible granules are obtained by mixing 75 parts by weight of a compound of the formula (I),
- 30
- | | | | | |
|----|---|---|---|-----------------------------|
| 10 | " | " | " | of calcium ligninsulfonate, |
| 5 | " | " | " | of sodium lauryl sulfate, |
| 3 | " | " | " | of polyvinyl alcohol and |
| 7 | " | " | " | of kaolin, |

grinding the mixture on a pinned disk mill, and granulating the powder in a fluidized bed by spraying on water as granulation liquid.

- 5 f) Water-dispersible granules are also obtained by homogenizing and precomminuting 25 part(s) by weight of a compound of the formula (I),

5	"	"	"	of sodium 2,2'-dinaphthyl-
				methane-6,6'-disulfonate,
10	2	"	"	of sodium oleoylmethyltaurate,
	1	"	"	of polyvinyl alcohol,
	17	"	"	calcium carbonate and
	50	"	"	of water

15 in a colloid mill, subsequently grinding the mixture in a bead mill, and atomizing the resulting suspension in a spray tower by means of a single-substance nozzle, and drying it.

- 20 g) Extruder granules are obtained by mixing 20 parts by weight of active substance, 3 parts by weight of sodium ligninsulfonate, 1 part by weight of carboxymethylcellulose and 76 parts by weight of kaolin, grinding the mixture, and moistening it with water. This mixture is extruded and subsequently dried in a stream of air.

25 C. Biological Examples

1. Pre-emergence effect on weeds

30 Seeds or rhizome pieces of monocotyledon and dicotyledon weed plants were placed in sandy loam soil in plastic pots and covered with soil. The compounds according to the invention which were formulated in the form of wettable powders or emulsion concentrates were then applied to the surface of the soil cover in the form of aqueous suspensions or emulsions at an application rate of 600 to 800 l of water/ha (converted), in various

dosages.

After the treatment, the pots were placed in a greenhouse and kept under good growth conditions for the weeds. After the test plants had emerged, the damage to the plants or the negative effect on the emergence was scored visually after a test period of 3 to 4 weeks by comparison with untreated controls. As shown by the score figures, the compounds according to the invention have a very good herbicidal pre-emergence action against a broad range of grass weeds and dicotyledon weeds. Compared with those sulfonylureas which correspond structurally to the salts, the salts according to the invention, for example those of Examples 1, 2, 3, 12, 13, 15, 29 and 52 of Table 1 show in some cases a considerably better herbicidal action in the case of monocotyledon weed species such as Avena, Alopecurus, Echinochloa, Digitaria, Setaria, Cyperus, Bromus or Sorghum or in the case of dicotyledon weed species such as Galium, Viola, Veronica, Lamium, Stellaria, Amaranthus, Sinapis, Pharbitis or Convolvulus.

2. Post-emergence effect on weeds

Seeds or rhizome pieces of monocotyledon and dicotyledon weeds were placed in sandy loam soil in plastic pots, covered with soil and grown in a greenhouse under good growth conditions. Three weeks after sowing, the test plants were treated in the three-leaf stage.

The compounds according to the invention which were formulated as wettable powders or as emulsion concentrates were sprayed in various dosages on the green parts of the plants at an application rate of 600 to 800 l of water/ha (converted) and, after the test plants had remained in the greenhouse for about 3 to 4 weeks under ideal growth conditions, the effect of the preparations was scored visually by comparison with untreated controls. The agents according to the invention also have a good herbicidal post-emergence action against a broad

range of economically important grass weeds and dicotyledon weeds. Compared with those sulfonylureas which correspond structurally to the salts, the salts according to the invention, for example those of Examples 1, 2, 3, 12, 13, 15, 29 and 52 of Table 1 show in some cases a considerably better herbicidal action in the case of monocotyledon weed species such as Avena, Alopecurus, Echinochloa, Digitaria, Setaria, Cyperus, Bromus or Sorghum or in the case of dicotyledon weed species such as Galium, Biola, Veronica, Lamium, Stellaria, Amaranthus, Sinapis, Pharbitis or Convolvulus.

3. Tolerance by crop plants

In further greenhouse experiments, seeds of a substantial number of crop plants and weeds were placed in sandy loam soil and covered with soil.

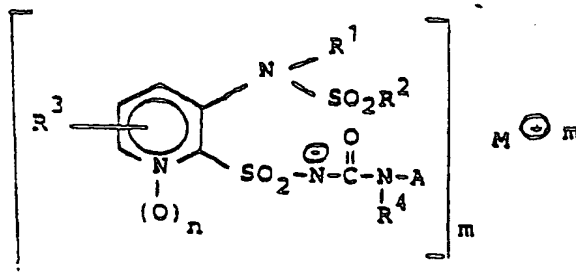
Some of the pots were treated immediately as described under 1., and the remaining pots were placed in a greenhouse until the plants had developed two to three true leaves and then sprayed with various dosages of the substances according to the invention, as described under 2.

Visual scoring four to five weeks after the application and after the plants had been in the greenhouse revealed that the compounds according to the invention did not inflict any damage to dicotyledon crops such as, for example, soya, cotton, oilseed rape, sugar beet and potatoes when used pre- and post-emergence, even when high dosages of active substance were used. Moreover, some substances also left Gramineae crops such as, for example, barley, wheat, rye, Sorghum species, maize or rice unharmed. The compounds of the formula (I) therefore have a high selectivity when used for controlling undesired plant growth in agricultural crops. The compounds of Examples 1, 2, 3, 12, 13, 15, 29 and 52 of Table 1 showed, for example, good selectivity properties

in cereal crops such as wheat and barley as well as in
rice, soya and maize.

PATENT CLAIMS

1. A compound of the formula (I)



in which

R^1 is H, (C_1-C_6) alkyl which is unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) alkoxy, (C_1-C_4) alkylthio, (C_1-C_4) alkylsulfinyl, (C_1-C_4) alkylsulfonyl, $[(C_1-C_4)$ alkoxy]carbonyl and CN, (C_3-C_6) -alkenyl which is unsubstituted or substituted by one or more halogen atoms, (C_3-C_6) alkynyl which is unsubstituted or substituted by one or more halogen atoms, (C_1-C_4) alkylsulfonyl which is unsubstituted or substituted by one or more halogen atoms, phenylsulfonyl where the phenyl radical is unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) alkyl and (C_1-C_4) alkoxy, (C_1-C_4) alkoxy or $[(C_1-C_4)$ -alkyl]carbonyl, which is unsubstituted or substituted by one or more halogen atoms,

R^2 is (C_1-C_4) alkyl which is unsubstituted or substituted by one or more halogen atoms, phenyl, where the phenyl radical is unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C_1-C_4) alkyl and (C_1-C_4) alkoxy, or is di- $[(C_1-C_4)$ alkyl]amino

or

R¹ and R² together are a chain of the formula
-(CH₂)_o- where the chain can additionally
be substituted by 1 to 4 (C₁-C₃)alkyl
radicals, and o is 3 or 4,

R³ is H, (C₁-C₄)alkyl, (C₁-C₃)haloalkyl,
halogen, NO₂, CN, (C₁-C₃)alkoxy, (C₁-C₃)-
haloalkoxy, (C₁-C₃)alkylthio, (C₁-C₃)-
alkoxy-(C₁-C₃)alkyl, [(C₁-C₃)alkoxy]car-
bonyl, (C₁-C₃)alkylamino, di-[(C₁-C₃)-
alkyl]amino, (C₁-C₃)alkylsulfinyl, (C₁-C₃)-
alkylsulfonyl, SO₂NR^aR^b or C(O)NR^aR^b,

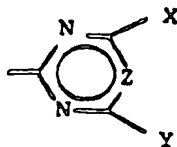
R^a and R^b independently of one another are H,
(C₁-C₃)alkyl, (C₃-C₆)alkenyl or propargyl,
or together are -(CH₂)₄-, -(CH₂)₅- or
-CH₂CH₂OCH₂CH₂-,

R⁴ is H or CH₃,

n is zero or 1,

m is 1 or 2,

A is a radical of the formula



X and Y independently of one another are H,
halogen, (C₁-C₃)alkyl, (C₁-C₃)alkoxy or
(C₁-C₃)alkylthio, the abovementioned
alkyl-containing radicals being unsub-
stituted or mono- or polysubstituted by
halogen or mono- or disubstituted by
(C₁-C₃)alkoxy or (C₁-C₃)alkylthio, further-
more a radical of the formula NR⁵R⁶,
(C₃-C₆)cycloalkyl, (C₂-C₄)alkenyl, (C₂-C₄)-
alkynyl, (C₃-C₄)alkenyloxy or (C₃-C₄)alky-
nyloxy,

Z is CH or N,

R⁵ and R⁶ independently of one another are H,

M

(C₁-C₃)alkyl or (C₃-C₄)alkenyl, and is an atom selected from the group comprising the alkali metals, alkaline earth metals or a group of the formula M1



5 in which R⁷, R⁸, R⁹ and R¹⁰ independently of one another are H, (C₁-C₁₂)alkyl, (C₃-C₆)alkenyl, (C₃-C₆)alkynyl, (C₃-C₈)-cycloalkyl or phenyl, the last 5 radicals mentioned in each case independently of
10 one another being unsubstituted or substituted by one or more radicals selected from the group comprising halogen, (C₁-C₄)alkyl, hydroxyl, (C₁-C₄)alkoxy, thio, (C₁-C₄)alkylthio, [(C₁-C₄)alkoxy]-
15 carbonyl and optionally substituted phenyl, or two of the radicals R⁷ to R¹⁰ together are a saturated or unsaturated chain of 3-7 carbon atoms, it being possible for 1-2 carbon atoms to be replaced
20 by atoms selected from the group comprising O, N or S and for the chain to be substituted by 1-3 (C₁-C₄)alkyl radicals, and the remaining two radicals have the abovementioned meanings of individual
25 radicals R⁷ to R¹⁰.

2. A compound as claimed in claim 1, wherein
R³, R^a, R^b and A are defined as in claim 1 and
n = zero,
R¹ is H, (C₁-C₄)alkyl which is unsubstituted
30 or substituted by one or more halogen atoms or by a radical selected from the group comprising (C₁-C₃)alkoxy, (C₁-C₃)-alkylthio, (C₁-C₃)alkylsulfonyl, [(C₁-C₄)-

- alkoxy]carbonyl and CN, or is (C₃-C₄)-alkenyl, (C₃-C₄)alkynyl, (C₁-C₄)alkylsulfonyl, (C₁-C₃)alkoxy or [(C₁-C₄)alkyl]carbonyl, and
- 5 R² is (C₁-C₄)alkyl which is unsubstituted or substituted by 1 to 3 radicals selected from the halogen group, or
- R¹ and R² together are a chain of the formula -(CH₂)_o-, in which o is 3 or 4.
- 10 3. A compound as claimed in claim 1 or 2, wherein
- R¹ is hydrogen, (C₁-C₄)alkyl or halo(C₁-C₄)alkyl,
- R² is (C₁-C₃)alkyl or halo(C₁-C₄)alkyl,
- R³ is H, (C₁-C₃)alkyl, (C₁-C₃)alkoxy, halogen or (C₁-C₃)alkylthio,
- 15 Z CH or N,
- X is (C₁-C₃)alkyl, (C₁-C₃)alkoxy, (C₁-C₃)haloalkyl, (C₁-C₃)haloalkoxy, (C₁-C₃)alkoxy-(C₁-C₃)alkyl or (C₁-C₃)alkoxy-(C₁-C₃)alkoxy, and
- Y is halogen, (C₁-C₃)alkyl, (C₁-C₃)alkoxy or (C₁-C₃)alkylthio, it being possible for the last
- 20 3 radicals mentioned to be monosubstituted or polysubstituted by halogen or monosubstituted or disubstituted by radicals selected from the group comprising (C₁-C₃)alkoxy and (C₁-C₃)alkylthio,
- 25 or a radical of the formula NR⁵R⁶ in which R⁵ and R⁶ have the abovementioned meaning, or (C₃-C₆)-cycloalkyl, (C₂-C₄)alkynyl, (C₃-C₄)alkenyloxy or (C₃-C₄)alkynyloxy.
- 30 4. A compound as claimed in one of claims 1 to 3, wherein X is (C₁-C₂)alkyl, (C₁-C₂)alkoxy, OCF₂H, CF₃ or OCH₂CF₃, and Y is (C₁-C₂)alkyl, (C₁-C₂)alkoxy, halogen or OCF₂H.
5. A compound as claimed in one of claims 1 to 4, wherein
- 35 M is Na, Li, K, Ca or a group of the formula M1

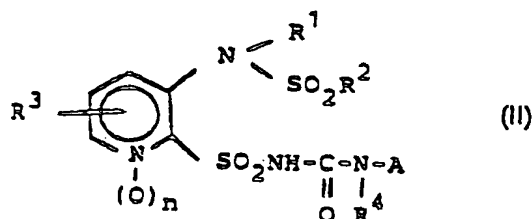


(M1)

in which R^7 , R^8 , R^9 and R^{10} independently of one another are H, (C_1-C_{12}) alkyl, (C_3-C_4) alkenyl, (C_3-C_4) alkynyl, (C_5-C_6) cycloalkyl, $[(C_1-C_4)$ alkoxy]-carbonyl- (C_1-C_4) alkyl, hydroxy- (C_1-C_4) alkyl, phenyl, benzyl, 1-phenylethyl or 2-phenylethyl, or

M is piperidine, pyrrolidine, morpholine or pyridine.

6. A process for the preparation of a compound of the formula (I) as claimed in claim 1, which comprises reacting a compound of the formula (II)



in which A, R^1 , R^2 , R^3 , R^4 and n are defined as in formula (I) as claimed in claim 1, with a base of the formula (IIIa) or (IIIb)



- 15 in which M and m are defined as in formula (I) as claimed in claim 1 and R^{11} is H or an aliphatic or aromatic organic radical and p is the number 2 in the event that m = 1 and the number 1 in the event that m = 2,

- 20 or, in the event that $R^{10} = H$, reacting a compound of the formula (II) with an amine of the formula (IV)



(IV)

in which R⁷ to R⁹ are defined as in formula (I) as claimed in claim 1.

- 5 7. The use of a compound of the formula (I) as claimed
 in one of claims 1 to 5 as a herbicide or plant
 growth regulator.
8. A herbicide and plant growth regulating composition,
 which comprises a compound of the formula (I) as
 claimed in one of claims 1 to 5 and conventional
 formulation auxiliaries.
- 10 9. A process for the selective or non-selective control
 of harmful plants or for regulating the growth of
 plants, which comprises applying one or more com-
 pounds of the formula (I) as claimed in one of
15 claims 1 to 5 to the plants, parts of the plants,
 seeds of the plants or the area under cultivation.

Office
Date

